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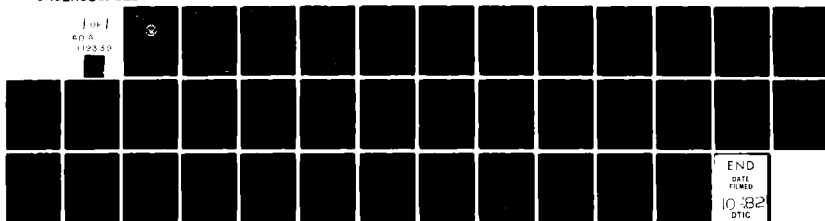
REAL GAS CONSIDERATIONS FOR DETERMINING PHYSICAL AND THERMODYNA--ETC(U)

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NAVAL POSTGRADUATE SCHOOL
Monterey, California



THESIS

**REAL GAS CONSIDERATIONS FOR DETERMINING
PHYSICAL AND THERMODYNAMIC PROPERTIES OF
GASSES INVOLVED IN THE PREDICTION OF THE
EFFECTS OF INTERNAL EXPLOSIONS**

by

Lewis Keith Athow

June 1982

Thesis Advisor:

R.A. Reinhardt

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
	AD-A119337	
4. TITLE (and Subtitle) Real Gas Considerations for Determining Physical and Thermodynamic Properties of Gases Involved in the Prediction of the Effects of Internal Explosions		5. TYPE OF REPORT & PERIOD COVERED Master's Thesis June 1982
7. AUTHOR(s) Lewis Keith Athow		6. PERFORMING ORG. REPORT NUMBER
8. PERFORMING ORGANIZATION NAME AND ADDRESS Naval Postgraduate School Monterey, California 93940		9. CONTRACT OR GRANT NUMBER(s)
11. CONTROLLING OFFICE NAME AND ADDRESS Naval Postgraduate School Monterey, California 93940		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		12. REPORT DATE June 1982
		13. NUMBER OF PAGES 37
		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Internal explosions; Real gas behavior; Vapor phase critical constants; Critical point; Critical constant estimation; Critical constants of metals; Equation of state; Magnesium; Aluminum; Magnesium Oxide		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) An investigation into the thermodynamic properties of gaseous magnesium, aluminum and their oxides has been conducted to determine the validity of assuming ideal-gas-law behavior in confined explosions when these metals are present. A number of vapor phase equations of state were tested and compared. The paucity of experimental and		

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Using the Dieterici equation, which proved satisfactory in terms of the two criteria given above, it was determined that deviation from ideal gas behavior was insignificant except in the case of metal oxides, which may have a determinable effect on the equilibrium conditions of the explosion products.

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Real Gas Considerations for Determining
Physical and Thermodynamic Properties of Gases
Involved in the Prediction of the Effects of Internal Explosions

by

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Lieutenant Commander, United States Navy
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Submitted in partial fulfillment of the
requirements for the degree of

MASTER OF SCIENCE IN PHYSICS

from the

NAVAL POSTGRADUATE SCHOOL
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ABSTRACT

An investigation into the thermodynamic properties of gaseous magnesium, aluminum and their oxides has been conducted to determine the validity of assuming ideal-gas-law behavior in confined explosions when these metals are present. A number of vapor phase equations of state were tested and compared. The paucity of experimental and rigorous theoretical data mandated the use of an equation of state that would provide realistic results and yet be tolerant of reasonable variations in the estimation of necessary parameters. The parameters selected are the vapor phase critical constants, which have been estimated from the theory of corresponding states or from partition function calculations.

Using the Dieterici equation, which proved satisfactory in terms of the two criteria given above, it was determined that deviation from ideal gas behavior was insignificant except in the case of metal oxides, which may have a determinable effect on the equilibrium conditions of the explosion products.

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TABLE OF SYMBOLS

a, b, c	Parameters used in various equations of state
f	Fugacity
f_f	Fugacity of the final state
f_i	Fugacity of the initial state
H	Enthalpy
P	Pressure
P_c	Critical Pressure
P_f	Pressure of the final state
P_i	Pressure of the initial state
P_r	Reduced pressure (P/P_c)
R	Gas law constant
T	Temperature
T_c	Critical Temperature
T_r	Reduced temperature (T/T_c)
V	Volume
V_c	Critical Volume
V_m	Molar Volume
V_r	Reduced Volume (V/V_c)
w	Fugacity coefficient (f/P)

I. INTRODUCTION

Recent investigations into predicting the effects of internal explosions have relied on computers for solutions to the many equations which are necessary to describe adequately the chemical and physical state of the products obtained from fuels consisting of carbon, hydrogen, nitrogen, and oxygen [Refs. 1 - 4]. Considerable work has also been done to describe the chemical equilibria of rapid internal combustions when metals such as aluminum and magnesium are present [Refs. 2 - 4]. Some of the more significant internal explosion work, INEX, sponsored by the Naval Weapons Center (NWC), China Lake, is being conducted simultaneously by researchers at the Naval Postgraduate School, Monterey, California, and at NWC, China Lake, California.

One particular simplifying assumption has remained in these recent investigations: the gaseous products of combustion obey the ideal gas law, $PV = nRT$. For the carbon, hydrogen, nitrogen, oxygen (C-H-N-O) systems, the assumption is quite valid, since these systems, when present as products of an internal explosion, are well removed from their critical point.

However, metal and metal oxide products of an internal explosion frequently exist as saturated or slightly superheated vapors, and may not behave in an ideal manner. The questionable ideality of the metal and metal oxide vapor warrants resolution in

order to describe or to predict more accurately the destructive capabilities of modern explosives containing selected metals [Ref. 4]. It is proposed that the assumption that the metal and metal oxide vapors obey the ideal gas law can either be fully justified or else be replaced by a more descriptive equation of state, and that the necessary thermodynamic properties of these vapors can be determined accordingly. It is intended that the results be directly applicable to the INEX work and generally applicable to any high temperature, high pressure systems.

II. NATURE OF THE PROBLEM

The assumption that a gas obeys the ideal gas law in a particular pressure, volume, temperature (P-V-T) region can be readily validated or repudiated if sufficient experimental data for the region is available for the gas. Unlike C-H-N-O systems, there is very little reliable data pertaining to the metal or metal oxides of interest in confined explosions. Since most explosives containing metals contain aluminum, magnesium, or both aluminum and magnesium, these metals and their oxides were investigated with respect to their behavior in the P-V-T conditions expected in an explosion occurring in a confined volume of air.

There are many equations of state which can be used to describe a gas under any particular conditions [Refs. 5 - 8]. Those equations which are more complex or which have more modifying parameters are generally more accurate and are valid over a larger P-V-T region. Hence, to qualify the ideal gas law assumption, it becomes a matter of arriving at one or more equations of state which will adequately describe the metal and metal oxides and then comparing the results of these equations to the results of the ideal gas law. When the appropriate equations of state are determined, other thermodynamic properties of the gas can be calculated for any particular conditions of P-V-T. The complexity of the appropriate equation of state will probably

require the use of numerical methods to calculate the thermodynamic properties of the gas. If so, it is intended that the calculation be adapted to a Hewlett-Packard 9845 desktop computer for solution.

III. PROCEDURE

A. THE SPECIES

Based on the work of other researchers, it has been determined that the gaseous metal/metal oxide species most likely to be encountered under the conditions of a confined explosion are aluminum (Al), aluminum oxide (Al_2O and AlO_2), magnesium (Mg), and magnesium oxide (MgO) [Refs. 3,4]. This specific investigation will involve these more common species, and methods of application to other species which may exist will be offered when feasible.

B. EQUATIONS OF STATE

Probably the most accurate equation of state is the virial equation [Refs. 6,7].

$$P = \frac{RT}{V_m} \left(1 + \frac{B(T)}{V_m} + \frac{C(T)}{V_m^2} + \dots \right) \quad (1)$$

It, however, has some serious disadvantages. It may involve a large number of terms, and the virial coefficients (B, C, \dots) are extremely difficult to determine. Considerable work has been done to determine the virial coefficients [Refs. 6,7], but it is still virtually impossible to arrive at more than the first three coefficients without rigorous mathematical models or without extensive experimental data [Refs. 6,7].

Other equations of state are simpler mathematically and have proven to be very descriptive of gaseous behavior in specified P-V-T regions [Refs. 5 - 10]. The simpler equations are van der Waals, Berthelot, Dieterici, Clausius, Wohl and Redlich-Kwong. Of these equations, the van der Waals equation is probably the one best known for its simplicity and reasonable accuracy. Most of these parametric equations of state require experimental data to determine the necessary parameters. Determining more parameters usually will enable a more accurate description of the gas. Without sufficient experimental vapor phase data from which to determine the necessary parameters for an equation of state, it becomes necessary to use other means to determine an equation of state. The statistical partition function has been used for simple molecules [Ref. 11], but the mathematical expression is rather complex and difficult to solve. Estimation techniques have been developed for many types of substances, but these estimation techniques generally work well only for organic compounds [Ref. 8]. Hence, the equations of state to be considered depend on the parameters which can be determined.

C. PARAMETERS FOR EQUATIONS OF STATE

Probably the most successful and simple method for estimating the parameters for an equation of state is using the Theorem of Corresponding States, along with the law of rectilinear diameters and the Clapeyron equation (see next paragraph). Although simpler estimation methods exist, such as the Guldberg relation which

states $T_c = 1.5T_b$ [Ref. 8], they are not particularly accurate for metals since metallic bonding has a significant influence on their molecular behavior [Refs. 12 - 14]. It is therefore desirable to compare the metals of interest to a known metal. Mercury is the only metal for which extensive vapor phase data is available.

The Theorem of Corresponding States indicates that substances which are at the same reduced temperature and pressure have equal reduced volumes. [Ref. 8]. The Law of Rectilinear Diameters states that the sum of the liquid and vapor densities of a substance is linear with pressure. This law has been shown to be valid for the metal liquid-vapor data that have been assembled [Refs. 12,14,15]. The Clapeyron equation, $dP/dT = \Delta H/(T V)$, is an exact thermodynamic result which applies to any phase change [Ref. 5].

The Theorem of Corresponding States, the Law of Rectilinear Diameters, and the Clapeyron equation have been used by several investigators to determine the critical constants of certain metals [Refs. 12,13,15,16]. The method is described quite well by Grosse [Ref. 12]. These determinations seem to be quite good based on the experimental data which have been obtained. McGonigal, Kirshenbaum, and Grosse used this method to estimate the critical constants of magnesium [Ref. 16]. Applying the theorem of corresponding states, it was claimed that a metal having a given entropy of vaporization would have the same reduced

temperature, T_r , as mercury at the same entropy of vaporization. The entropy of vaporization for magnesium at the normal boiling point (1390K) is 22.1 cal/gram-atom-degree, which corresponds to a reduced temperature of 0.36 on the entropy of vaporization vs. reduced temperature curve for mercury. Hence, the critical temperature is estimated to be 1390K/0.36 or 3850K, with an arbitrary error of $\pm 10\%$.

Experimental density data were obtained by McGonigal to determine the critical density of magnesium. An average of the vapor and liquid densities plotted vs. temperature is linear and passes through the critical point in accordance with the law of rectilinear diameter. From their data, the critical density of magnesium was estimated to be 0.41 ± 0.05 g/cm³ for a critical volume of 59 ± 8.0 cm³/mole.

Clausius integrated the Clapeyron equation:

$$\ln P = - \frac{\Delta H}{R} \frac{1}{T} + \text{constant}$$

where ΔH in this case represents the difference between the enthalpy of the saturated vapor and the enthalpy of the liquid (this is the Clausius-Clapeyron equation). The assumptions used for the integration were: (1) negligible liquid volume, (2) constant ΔH , and (3) ideal gas behavior of the vapor. None of these assumptions holds near the critical point. Nonetheless, it has been observed [Ref. 17] that $\ln P$ vs. $1/T$ plots are linear

from the low pressure region up to the critical point, a result of compensating effects from the three approximations.

In a similar manner, Goodwin, Johnson, and Wright [Ref. 15] determined the critical constants of aluminum. Leu, Ma, and Eyring used a partition function calculation to estimate the critical constants of magnesium oxide [Ref. 11]. The critical constant estimates for aluminum, magnesium, and magnesium oxide are given below.

<u>SUBSTANCE</u>	<u>T_c (K)</u>	<u>P_c (atm)</u>	<u>V_c (dm³/mole)</u>	<u>REFERENCE</u>
Mg	3850	1730	.0590	16
MgO	5950	33.47	.2095	11
Al	8000	4107	.0485	15

It is considered meaningless to estimate additional parameters: without supportive experimental data any additional parameters would be estimated from the critical constants [Ref. 8].

D. AN APPROPRIATE EQUATION

With three parameters available, there are a number of very good equations of state that can be considered for utilization [Refs. 5,8,9]. The principal equations of consideration are listed in Appendix A. To ensure the P_c-V_c isotherm passes through the critical point, it is desirable to use the equations of state in their reduced form, where the parameters, a and b, and the gas-law constant, R, are eliminated by solving the equations in terms of P_r, V_r, T_r, V, and T. Introducing the critical constants,

the equations can be solved in terms of P_c , V_c , T_c , P , V , and T . This method is described in detail by Sterbacek [Ref. 8]. Many of the popular equations have been compared in detail [Ref. 9]. An independent comparison of some of these equations is shown in Figures 1 through 4. In Figures 1 and 2, the equations were used in their parametric, non-reduced form. It is seen that the critical isotherm passes near the estimated critical point, but not through it. In Figures 3 and 4, the reduced equations always pass through the estimated critical point due to the conditions required of a critical isotherm [Ref. 8]: (1) At the critical point, the slope of the isotherm is zero and (2) The critical point is an inflection point for the critical isotherm. Essentially, using the above conditions with the three critical constants, we are adding a third parameter to a two parameter equation of state. Further comparison of the equations of state P-V diagrams of Figures 1 and 3 shows that for temperatures somewhat below the critical temperature the pressure becomes negative [Ref. 9], whereas the Gibbs phase rule [Ref. 5] requires that the isotherms be horizontal inside the saturated vapor-liquid envelope below the critical point [Refs. 5,8]. Note in Figure 4 that the Berthelot and van der Waals equations superimpose exactly. The two equations differ by a factor of T_r in their second terms; this factor is unity at the critical temperature.

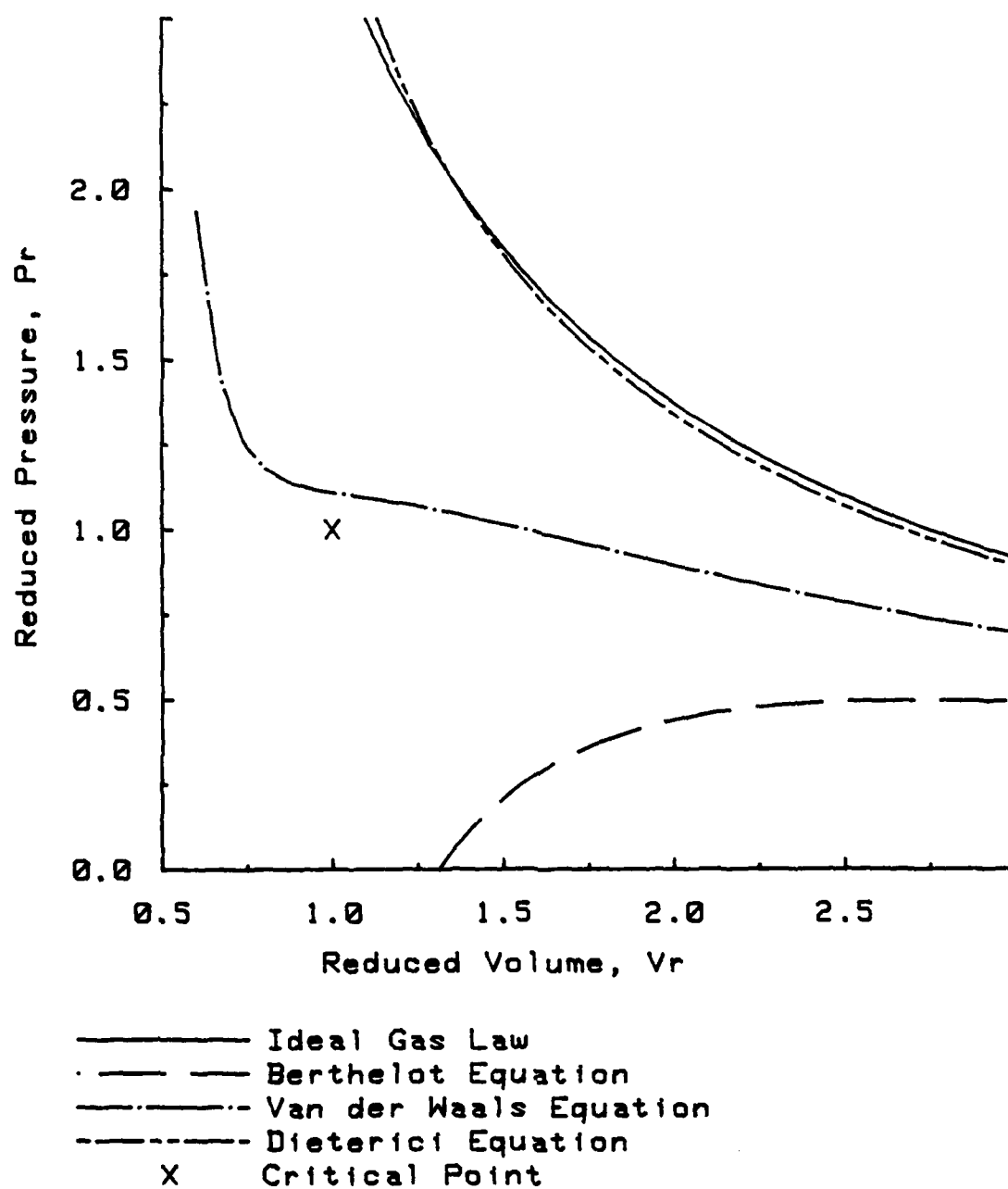


FIGURE 1. P-V Diagram.
Parametric Equations of State
Temperature = $.8T_c$

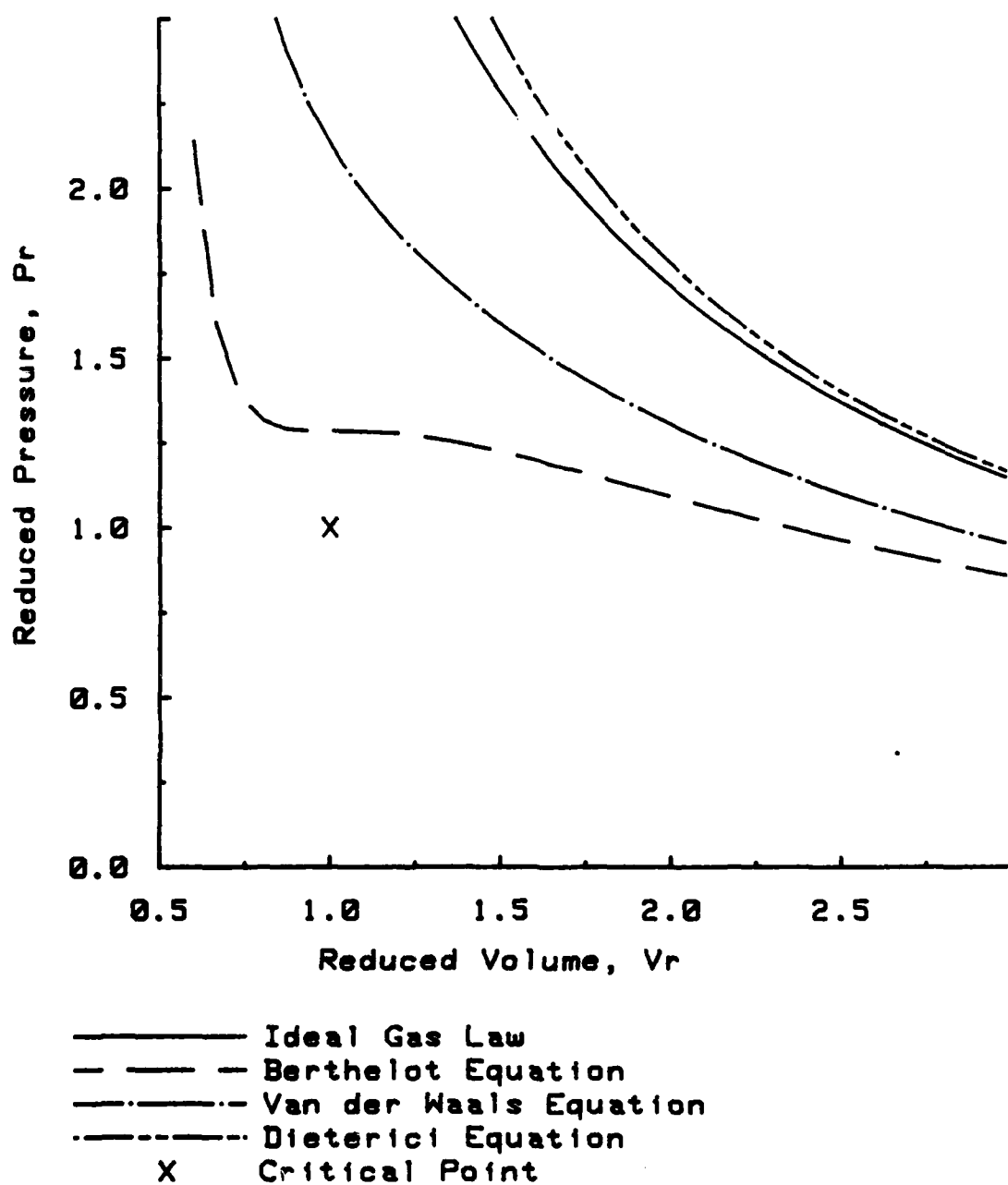


FIGURE 2. P-V Diagram.
Parametric Equations of State
Temperature = $1T_c$

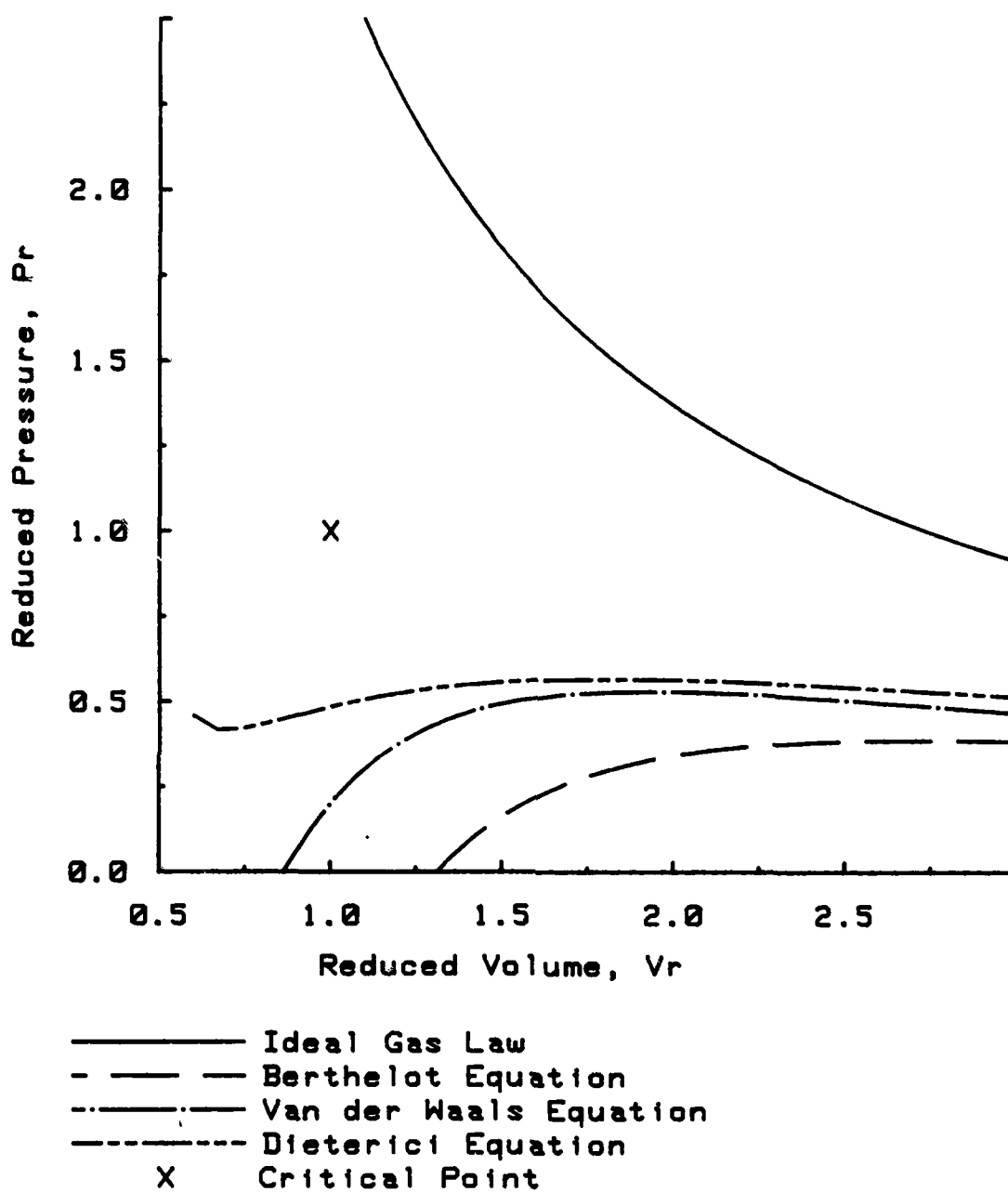


FIGURE 3. P-V Diagram.
Reduced Equations of State
Temperature = $.8T_c$

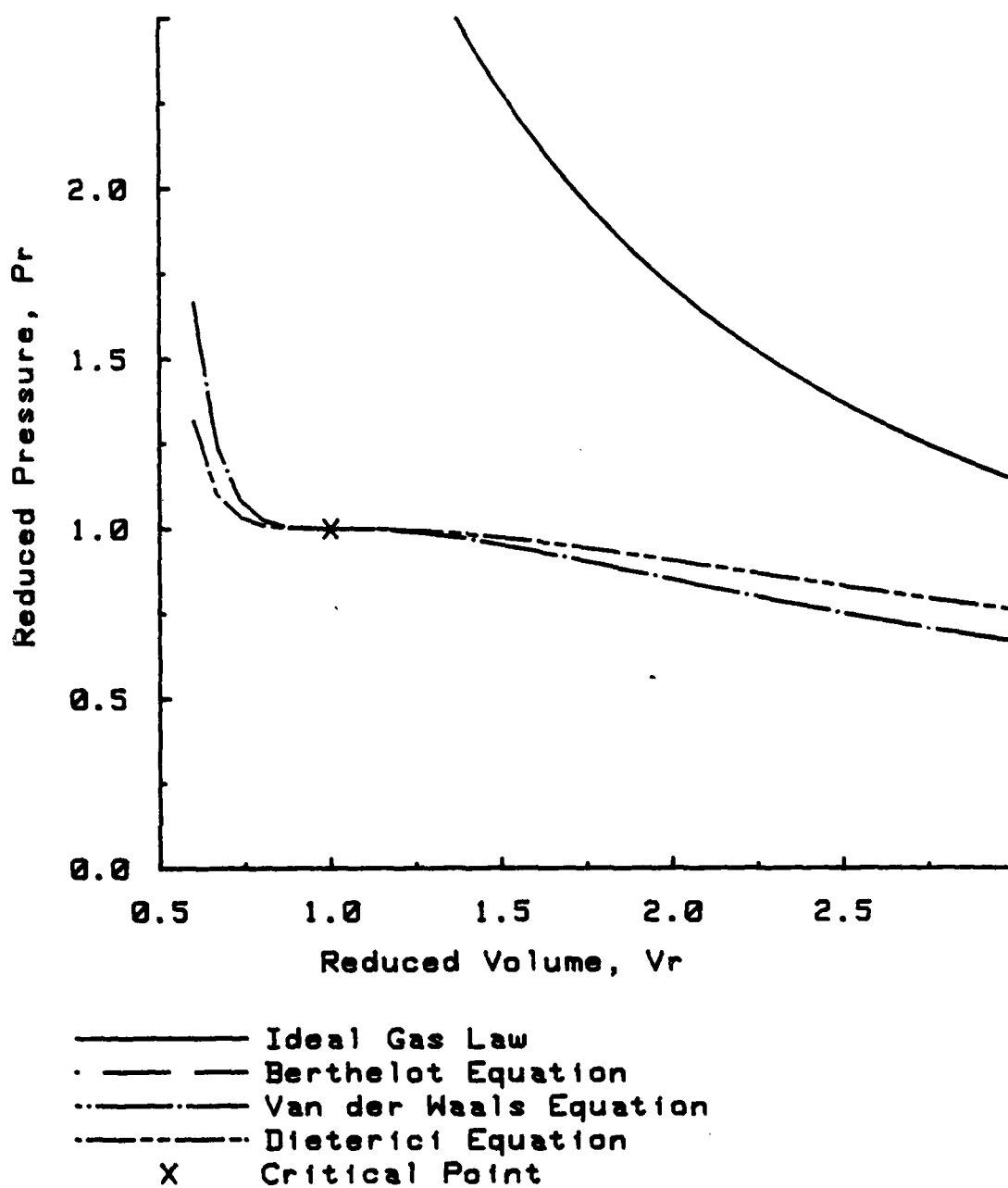


FIGURE 4. P-V Diagram.
 Reduced Equations of State
 Temperature = $1T_c$

The Dieterici equation isotherms, however, do remain horizontal or nearly horizontal at temperatures significantly below the critical temperature. This fact makes the Dieterici equation preferable to others when critical constant estimation techniques are employed.

Since the critical constants have only been estimated, it is reasonable to assume an error of ± 10 to 20%. Say, for example, that the critical temperature of aluminum, estimated to be 8000K is actually 6700K. Aluminum vapor at 6800K and at its critical volume, V_c , would then be a superheated gas at a pressure greater than its critical pressure, P_c . Using the Van der Waals equation to calculate the pressure at 6800K and at the critical volume using the estimated T_c of 8000K would indicate a pressure near zero. The Dieterici equation, on the other hand, using the same estimated T_c of 8000K would show a pressure on the same order as the actual critical pressure for a temperature of 6800K. This demonstrates the greater tolerance that the Dieterici equation has for the inaccuracy of the critical constant estimation. The other two and three parameter equations of state do not share this property. The major disadvantage of the Dieterici equation has been its exponential form, but a computer can be used effectively to overcome this disadvantage.

Once the Dieterici equation of state is selected as the one best suited for use, based on the estimate of critical constants, the P-V-T relationship it provides can be examined in the regime

anticipated by previous internal explosion research. The results of this examination should provide the information necessary to determine the validity of the ideal-gas-law assumption.

E. THERMODYNAMIC PROPERTIES

Certainly a big factor in using the ideal gas law instead of a more descriptive equation of state is the ease of solving the integrations to determine thermodynamic properties such as fugacity (or activity). Using the more complex equations of state makes the solutions more difficult; a computer and numerical methods facilitate the solutions and permit the use of more complicated integrands.

An expression leading to determination of the fugacity and the chemical activity can easily be written [Ref. 5]. The solution can be determined numerically.

For a real gas,

$$\int_{P_i}^{P_f} V_m dP = RT \ln \frac{f_f}{f_i} \quad (2)$$

For an ideal gas,

$$\int_{P_i}^{P_f} V_m dP = RT \ln \frac{P_f}{P_i} \quad (3)$$

Subtracting (3) from (2),

$$\ln \frac{f_f/P_f}{f_i/P_i} = \frac{1}{RT} \int_{P_i}^{P_f} (v_m^{\text{real}} - v_m^{\text{ideal}}) dP \quad (4)$$

If P_i approaches 0, then f_i approaches 0 and f_i approaches P_i or $f_i/P_i = 1$. Since

$$v_m^{\text{ideal}} = \frac{RT}{P} \text{ and } v_m^{\text{real}} = \frac{RTZ}{P} \text{ then}$$

substituting into (4) and dropping the subscript f gives

$$\ln \frac{f}{P} = \int_0^P \frac{Z-1}{P} dP$$

or

$$f = P e^{\int_0^P \frac{Z-1}{P} dP}$$

For the Dieterici equation,

$$P = \frac{P_c T_r e^{2/(T_r V_r)} e^{-2/(T_r V_r)}}{2V_r - 1}$$

or the compressibility factor, Z , is

$$Z = \frac{P_c T_c^2 e^{-2/(T_r V_r)}}{T(2V_r - 1)} \frac{V}{RT}$$

and the fugacity, f , is

$$f = P \exp \left\{ \int_0^P \frac{P_c V T_c^2 e^{-2/(T_r V_r)}}{P R T^2 (2V_r - 1)} - \frac{1}{P} dP \right\}$$

Since $f = wP$ where w is the fugacity coefficient, the exponential term in the above equation is the fugacity coefficient.

Similarly, other thermodynamic properties can be determined. Calculating thermodynamic properties as outlined above should more accurately describe the behavior of magnesium oxide. However, the behavior of magnesium oxide in a mixture of many gasses is somewhat uncertain. Ideally, the fugacity in the mixture could be determined by $f = xf^*$ where x is the mole fraction of MgO in the mixture and f^* is the fugacity of pure MgO at the temperature and total pressure of the mixture. A method of determining psuedo-critical constants for a gaseous mixture has been used [Ref. 18], but

primarily for binary mixtures, whose component critical constants do not differ greatly. Again, without experimental data, the exact behavior is uncertain. It may be that using the ideal mixture relationship is the best approach, but additional investigation and verification is most certainly necessary.

IV. ALTERNATE SOLUTIONS

The intent of this investigation was to validate the ideal gas law assumption for internal explosions. In doing this validation, it was necessary to use untested estimates of the critical constants for the species of interest. To produce more accurate results or to verify the critical constants it would be necessary to obtain extensive experimental P-V-T data for the species. Due to the extremely high temperature and pressures necessary, this method is not currently feasible.

Alternately, a rigorous mathematical model, such as a partition function could be developed as was done for MgO [Ref. 11], but the results obtained therefrom would realistically need to be verified with some experimental data - again such verification is not currently feasible.

V. CONCLUSIONS

A. REAL VS. IDEAL GAS CALCULATIONS

From previously published reports [Refs. 1,3,4], temperatures less than 5000K and pressures less than 200 atm can be expected for the internal explosions evaluated in other research. Figures 5, 6 and 7 show a comparison of the Dieterici equation and the ideal gas law for this P-T range for a density of 0 to 10kg/m³. The curves for Al, Mg, and MgO are shown. It can be seen that in the P-V-T region of interest, the deviation from ideality is quite small except in the case of MgO, where the deviation is rather significant. For the expected amounts of MgO to be produced [Ref. 3], the overall contribution to the total overpressure resulting from the explosion is considered to be quite small. However, assuming reasonable accuracy for the critical constants used for MgO, the actual concentration of MgO in the product mixture may significantly affect the solution of the equilibrium equations used in the INEX computer program which assumes ideal behavior. Additionally, it can be anticipated that the oxides of aluminum may behave in a manner similar to MgO. If this is the case, the overall impact on the system of explosion products could be quite significant.

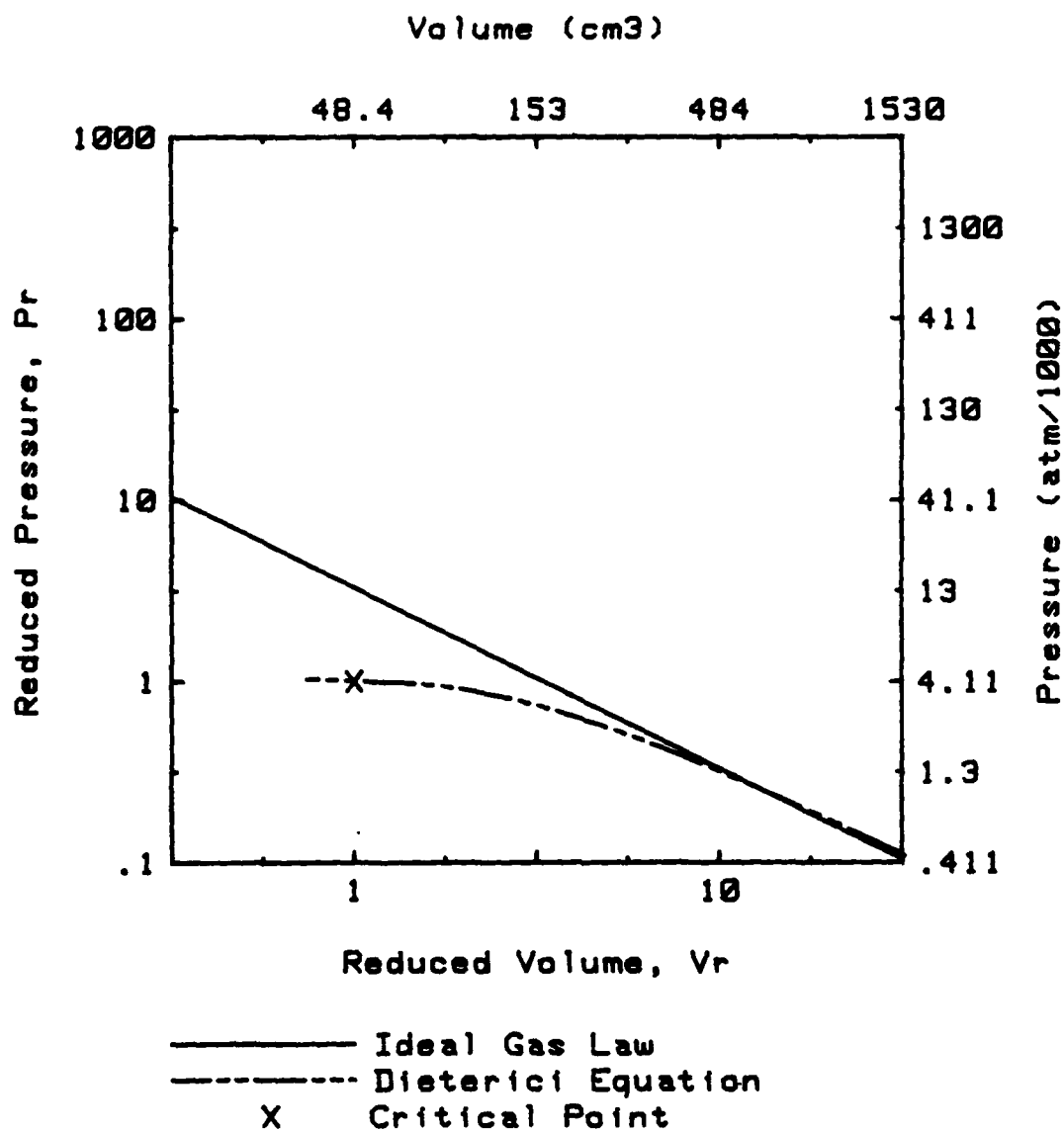


FIGURE 5. P-V Diagram. (Log Scale)
 Equations of State
 Temperature = T_c (8000 K)
 ALUMINUM

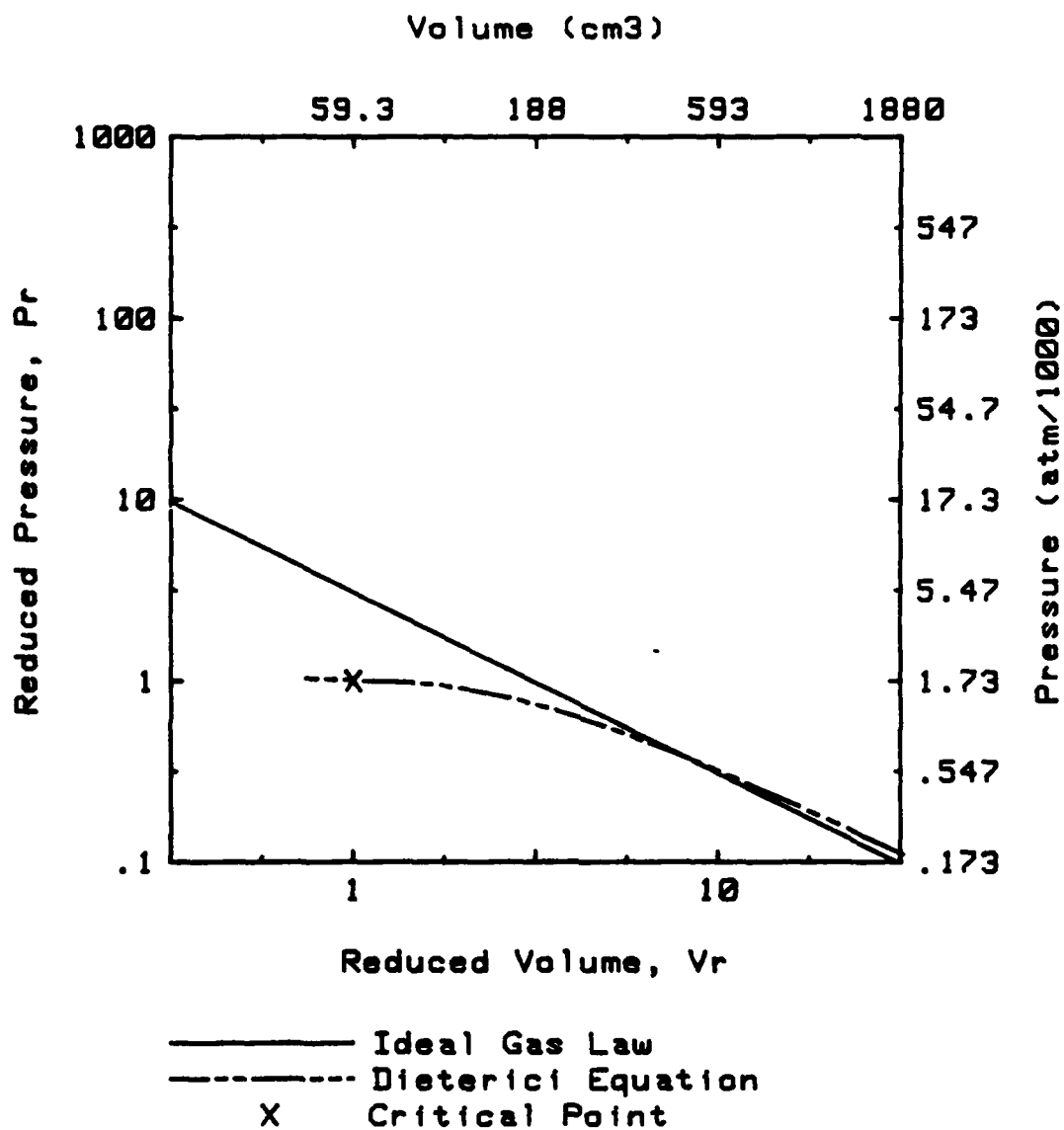


FIGURE 6. P-V Diagram. (Log Scale)
 Equations of State
 Temperature = T_c (3850 K)
 MAGNESIUM

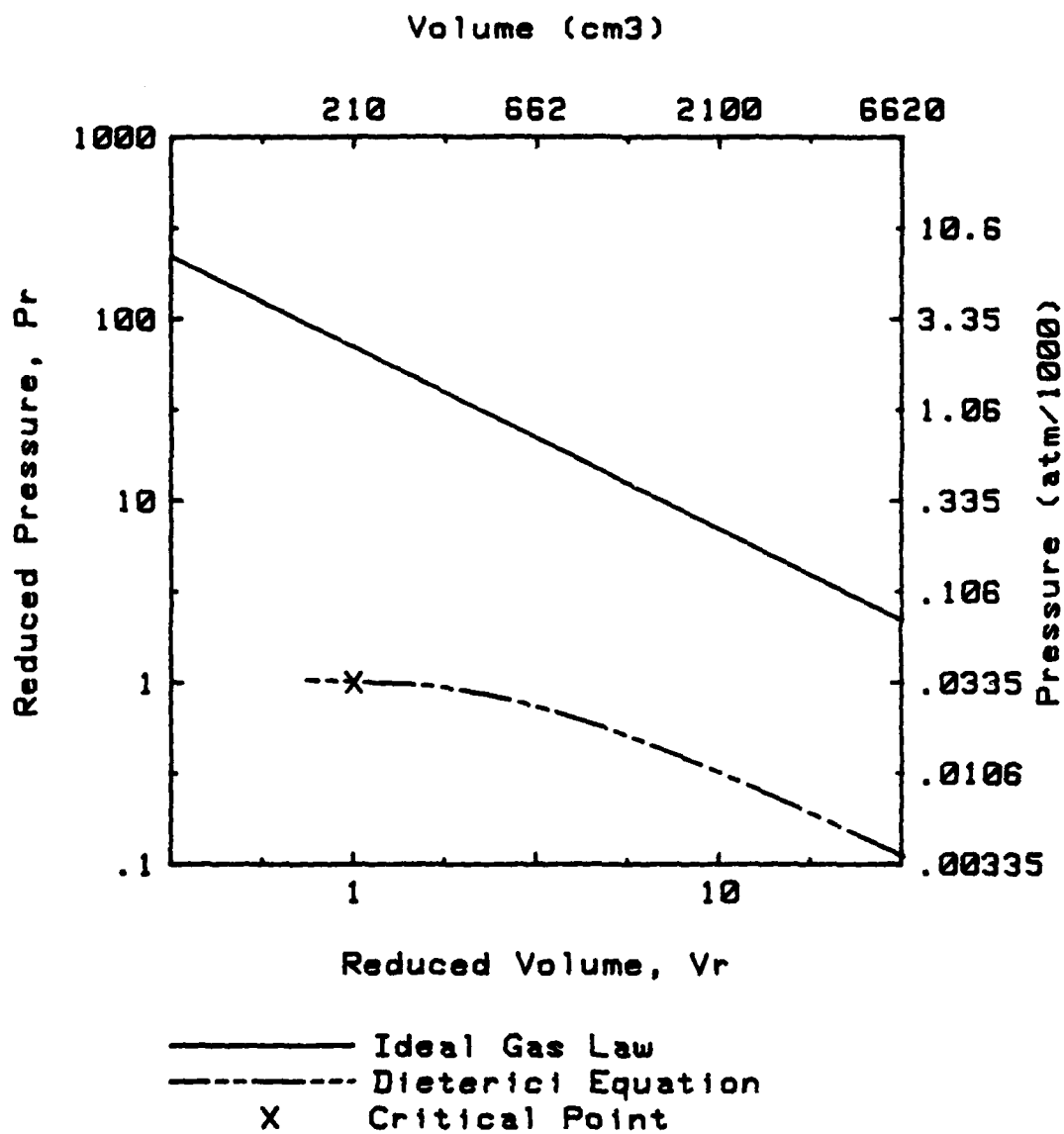


FIGURE 7. P-V Diagram. (Log Scale)
 Equations of State
 Temperature = T_c (5950 K)
 MAGNESIUM OXIDE

B. DIETERICI EQUATION OF STATE

It is noted that of the many popular two or three parameter equations of state, the Dieterici equation provides a reasonable tolerance for the inaccuracies that may exist in the estimation of the critical parameters. This tolerance is desirable when the P-V-T region near the vapor-liquid critical point is investigated.

Using results of the Dieterici equation applied to the P-V-T region anticipated for internal explosions, it is noted that there is no significant departure from ideal behavior except in the case of MgO, where the difference is significant and may have an important effect on equilibrium calculations.

C. APPLICATIONS

The Dieterici equation of state expressions for the fugacity and fugacity coefficient are not easily solved directly. However, in view of the significant deviation from ideal behavior exhibited by the magnesium oxide vapor, application of the Dieterici equation to a computer solution is considered to be of significant value for future internal explosion research.

APPENDIX A - SELECTED EQUATIONS OF STATE

Some of the more common two and three parameter equations of state are listed below.

1. Van der Waals:
$$P = \frac{RT}{V_m - b} - \frac{a}{V_m^2}$$
2. Berthelot:
$$P = \frac{RT}{V_m - b} - \frac{a}{TV_m^2}$$
3. Clausius:
$$P = \frac{RT}{V_m - b} - \frac{a}{T(V_m + c)^2}$$
4. Redlich-Kwong:
$$P = \frac{RT}{V_m - b} - \frac{a}{T^{0.5}V_m(V_m + b)}$$
5. Wohl:
$$P = \frac{RT}{V_m - b} - \frac{a}{TV_m(V_m - b)} + \frac{c}{T^2V_m^3}$$
6. Dieterici:
$$P = \frac{RT}{V_m - b} \exp(-a/(RTV_m))$$

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